Group-Theoretical Analysis of Second Harmonic Generation at (110) and (111) Surfaces of Antiferromagnets

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Extending our previous work we classify the nonlinear magneto-optical response at low index surfaces of fcc antiferromagnets, such as NiO. Antiferromagnetic bilayers are discussed here as models for the termination of bulk antiferromagnets.

I. INTRODUCTION

Nonlinear optics has been proven to be very useful for the investigation of ferromagnetism at surfaces due to its enhanced sensitivity to twodimensional *ferromagnetism* [1]. The magnetic effects are usually much stronger than in linear optics (rotations up to 90°, pronounced spin polarized quantum well state oscillations [2–4], magnetic contrasts close to 100%) [4–7]. Recently, Second Harmonic Generation (SHG) has been successfully applied to probe *antiferromagnetism* (visualization of bulk AF domains [8–10]). The potential of SHG to study the surface antiferromagnetism has been announced in Ref. [11] extensively discussed in our previous paper [12].

The practical importance of studies in this field follows from the applications of antiferromagnetic (AF) oxide layers in devices such as those based on TMR (tunneling magnetoresistance), where a trilayer structure is commonly used. The central layer of TMR devices consists of an oxide sandwiched between a soft and a hard magnetic layer. For these technological applications it is necessary to develop a technique to study buried oxide interfaces. Such a technique can be SHG. One of the promising materials for the mentioned devices is NiO. However, to the best of our knowledge, the understanding of its detailed spin structure is scarce - even the spin orientation on the ferromagnetically ordered (111) surfaces is not known.

Our recent paper [12] presented an extensive study of the nonlinear electrical susceptibility tensor $\chi_{el}^{(2\omega)}$ (the source for SHG within the electrical dipole approximation), mostly for monolayer structures. It has been proven there that the spin structure of an antiferromagnetic monolayer can be detected by means of SHG. Also the possibility of antiferromagnetic surface domain imaging has been presented for the first time. As it was mentioned in this previous work, bilayer spin structures are enough to account for the symmetry of a surface of a cubic antiferromagnet. Here we present an extention of that work to the (110) and (111) bilayer structures, thus completing our group theoretical analysis of low Miller-index antiferromagnetic surfaces.

II. RESULTS

We follow exactly the group theoretical method described in Ref. [12]. At this point it is necessary to define the notions of "phase" and "configuration", used henceforth to classify our results. "Phase" describes the magnetic phase of the material, i.e. paramagnetic, ferromagnetic, or AF. Secondly, the word "configuration" is reserved for the description of the magnetic ordering of the surface. It describes the various possibilities of the spin ordering, which are different in the sense of topology. We describe AF configurations, denoted by little letters a) to l), as well as several ferromagnetic configurations, denoted as "ferro1", "ferro2", etc. The number of possible configurations varies depending on the surface orientation. All the analysis concerns collinear antiferromagnets, with one easy axis.

The tables show the allowed tensor elements for each configuration. The tables also contain the information on the parity of the nonvanishing tensor elements: the odd ones are printed in boldface. In some situations an even tensor element (shown in lightface) is equal to an odd element (shown in boldface), this means that this pair of tensor elements is equal in the domain which is depicted on the corresponding figure, but they are of opposite sign in the other domain. The parity of the elements has been checked in the operations 2_z, 4_z, and in the operation connecting mirror-domains to each other (for the definition of the mirror-domain structure see Ref. [12]). The domain operation(s) on which the parity depends is (are), if applicable, also displayed in the tables. If two or more domain operations have the same effect, we display all of them together. To make the tables shorter and more easily readable some domain operations (and the corresponding parity information for the tensor elements) are not displayed, namely those that can be created by a superposition of the displayed domain operations. We also do not address the parity of tensor elements in the 6_z nor 3_z operations for (111) surfaces nor any other operation that "splits" tensor elements, although these operations

also lead to a domain structure [13]. As has been discussed in Ref. [12] it is possible to define a parity of the tensor elements for the 3_z and 6_z operations, however the tensor elements then undergo more complicated changes. The situations where the parity of the tensor elements is too complicated to be displayed in the tables are indicated by a hyphen in the column "domain operation". For the paramagnetic phase, where no domains exist, we display the hyphen as well. For some configurations, none of the operations leads to a domain structure - in those configurations we display the information "one domain". The reader is referred to Ref. [12] for the particularities of the parity check.

A. (110) bilayer

The previously described AF configurations of the (001) monolayer most commonly get split into two different configurations when a bilayer structure is considered. For the (110) bilayer it is not the case - only two of twelve AF configurations get split in this way, thus one obtains 14 AF configurations of the (110) bilayer. Describing the results of our analysis we use the nomenclature of our previous article, i.e. the antiferromagnetic configurations are labeled by small letters. Only the four configurations that result from splitting of the two configurations of the monolayer structure are labeled by small letters with subscripts that carry the information about how they have been constructed from the (110) monolayer. For configurations with subscript "a" the lower layer is constructed by translation of the topmost layer by vector (0.5a, 0.5b), where a and b are interatomic distances *within* the (110) plane along x and y axes, respectively. For configurations with subsript "b" the vector of translation is (-0.5a, 0.5b). This corresponds to the way we constructed the (001) bilayers in [12].

The configurations of the (110) monolayer structure are depicted in Fig. 1, and the way the bilayer is constructed is depicted in Fig. 2. The tensor elements are presented in Table I. In general, we can observe five types of response. However, the possibility to distinguish AF configurations is not much improved compared to the (110) monolayer. Even the possibility to detect the magnetic phase of the surface is not evident.

As for the (001) surface [12], there is no difference in SHG signal between the monolayer and bilayer for the paramagnetic and ferromagnetic phases. For most AF configurations, however (confs. a), b), c), e), f_a, f_b), g), h), j), k), and l)) such a difference is present due to a lower symmetry of the bilayer.

B. (111) bilayer

In order to be consistent with our previous work [12] we keep the same configuration names as in this earlier paper. That is why, for example, conf. b) is not present here. The spin configurations of the (111) bilayer are constructed from the configurations of the (111) surface of our previous work in the way that the spin structure in the second atomic layer is the same as in the topmost layer, but shifted accordingly to form a hcp structure. Taking into account the spin structure of the second layer causes all the AF configurations to split, thus one obtains 10 AF configurations of the (111) bilayer. The configurations are labeled by small letters (indicating their "parent" configuration) with subscript "a" if the mentioned shifting is along the positive x axis, and "b" if the shifting is along the negative S_{xy} axis.

The configurations of the (111) monolayer are depicted in Fig. 3 and the construction of the bilayer is depicted in Fig. 4. The corresponding tensor elements are displayed in Tab. II. The results are identical to those of our previous work [12], where the second layer of the (111) surface was treated as nonmagnetic. This means that the spin spin structure of the second layer does not play any role for SHG, however the presence of the atoms in the second layer does.

III. CONCLUSION

From our results follows that SHG can probe maximally two atomic layers of the surface of cubic two sublattice antiferromagnets, and only one of the paramagnetic or ferromagnetic surface. For the (111) surface, the *spin* structure of the second layer does not have any influence on SHG, i.e. it does not matter from the group-theoretical point of view if the investigated surface is a termination of a bulk antiferromagnet or a monolayer grown on a nonmagnetic substrate. However, these two situations can be very different from the band-theoretical point of view.

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[13] The notion of the parity operation requires that the original situation is restored after applying the operation at most twice. Obviously, 2_z is such an operation, and for certain configurations 4_z satisfies this criterion as well (where 2_z is just a symmetry operation). However, neither 3_z nor 6_z have this property - they must be applied at least 3 times to restore the original situation. Concerning the tensor elements, for 2_z and mirror-operations a given tensor element may change its sign or not. Upon the application of the 4_z operation it may become another tensor element (i.e. its x indices become y and vice versa) while its sign can change or remain unchanged. Upon the 3_z operation, however, a single tensor element gets "split" into several tensor elements, and the notion of the conservation (or not) of the sign of the tensor element looses its sense. The tensor elements are described in the cartesian coordinate system, where quarters are the elementary entities. Rotation by an angle other than 90° and its multiples cannot be described as interchanging the axes and a possible modification of their signs. However, it is possible to treat the parity of tensor elements in the 3_z and 6_z operation in a more complicated way.

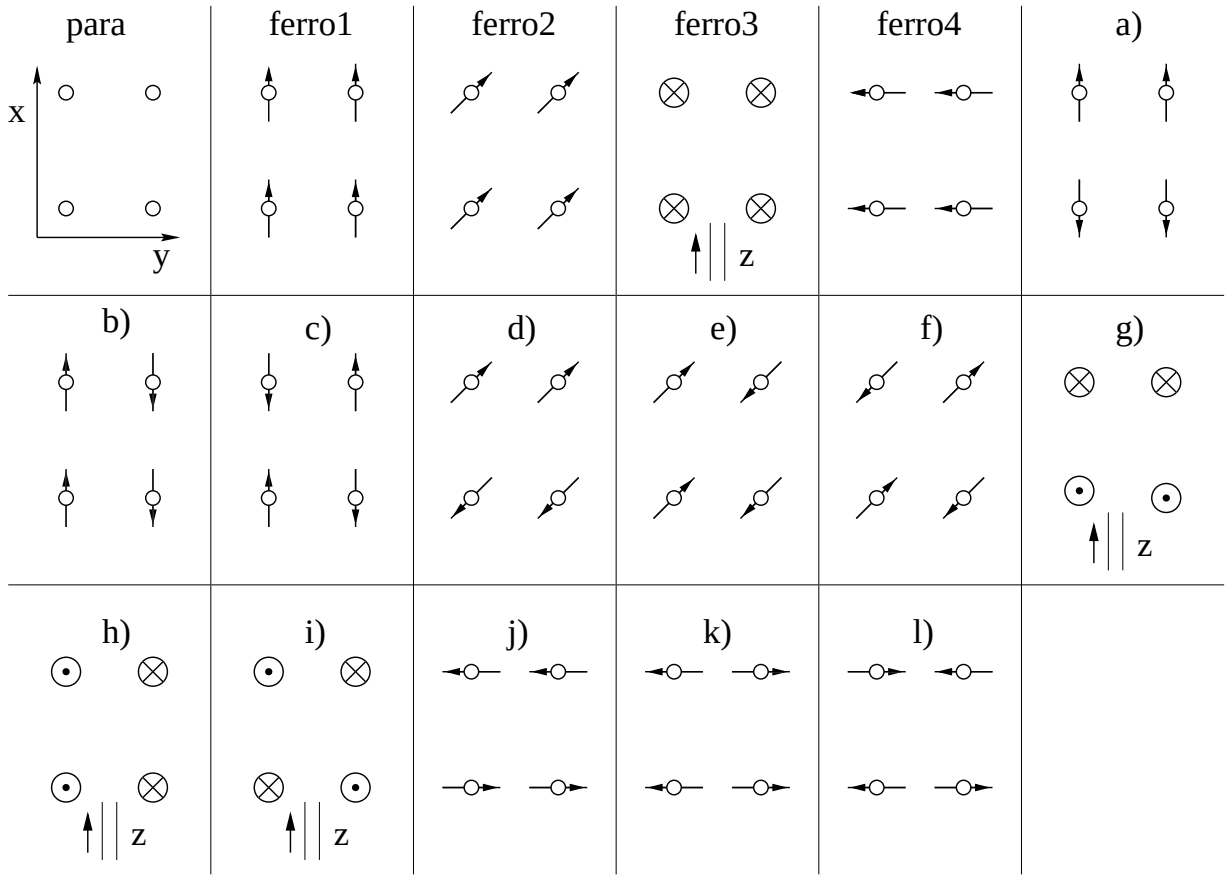

FIG. 1. Spin configurations of an fcc (110) monolayer. Except for confs. "ferro3", g), h), and i), the arrows always indicate in-plane directions of the spins. In confs. "ferro3", g), h), and i) $\odot$ ($\otimes$) denote spins pointing along the positive (negative) z-direction, respectively.

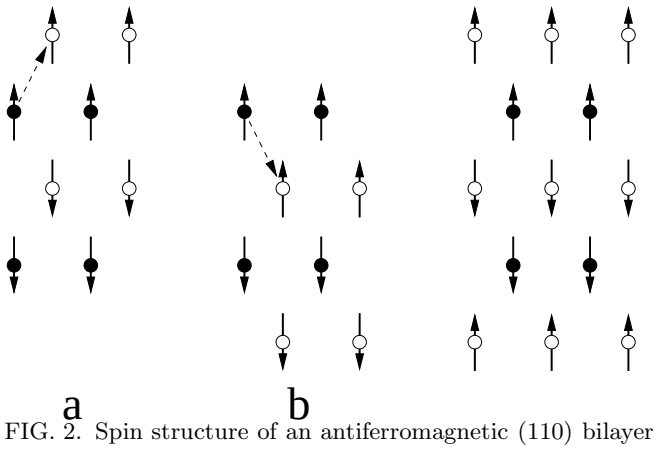

FIG. 2. Spin structure of an antiferromagnetic (110) bilayer constructed from a shift of the monolayer, where two different shiftings are applied. Filled (empty) circles represent the topmost (second) layer. The rightmost panel shows the conventional unit cell for the resulting bilayer structure. Here, conf. a) of the (110) surface serves as an example.

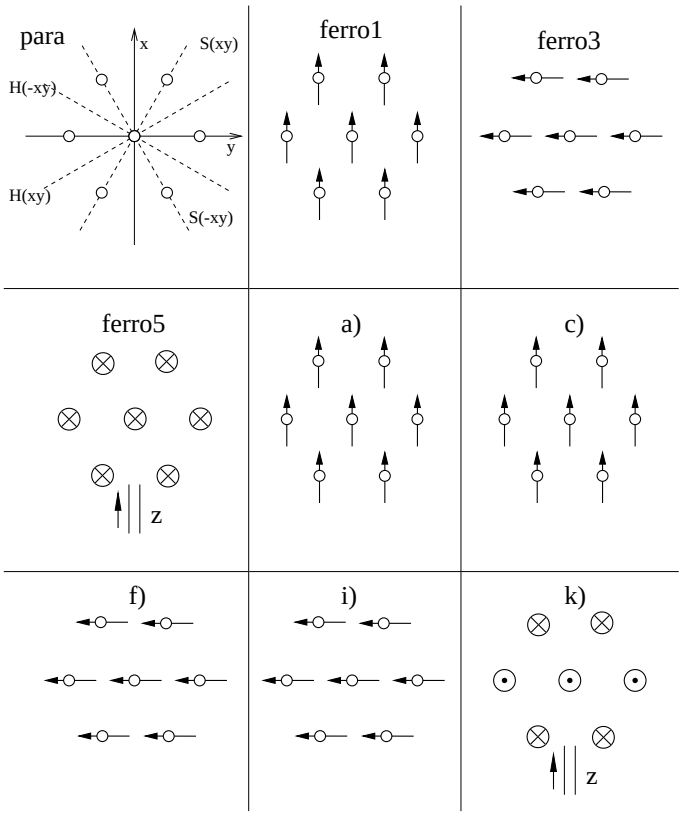

FIG. 3. Spin configurations of an fcc (111) monolayer. Except for confs. "ferro5", k), l), and m), the arrows always indicate in-plane directions of the spins. In confs. "ferro5" and k) $\odot$ ($\otimes$) denote spins pointing along the positive (negative) z-direction, respectively.

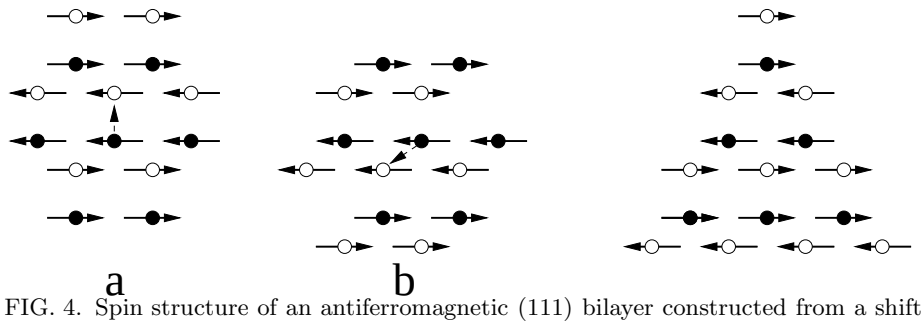

FIG. 4. Spin structure of an antiferromagnetic (111) bilayer constructed from a shift of the monolayer, where two different shiftings are applied. Filled (empty) circles represent the topmost (second) layer. Here, conf. f) of the (111) monolayer serves as an example. The rightmost panel displays the conventional unit cell for the resulting bilayer structure of conf. f_a).

TABLE I. Nonvanishing elements of $\chi_{el}^{(2\omega)}$ for all spin configurations of the (110) surface of a fcc lattice. We denote $\chi_{ijk}^{(2\omega)}$ by ijk. Odd elements are in bold if a parity operation exists. The configurations are depicted in Fig. 1.

conf.	point group	symmetry operations	domain operation	non-vanishing tensor elements
para	mm2	$1, 2_z, \overline{2}_x, \overline{2}_y$	-	xxz = xzx, yyz = yzy, zxx, zyy, zzz
ferro1	m	$1, \overline{2}_x$	$2_z, \overline{2}_y$	xzx = xxz, **xxy** = **xyx**, **yxx**, **yyy**, **yzz**, yyz = yzy, zxx, zyy, zzz, **zyz** = **zzy**
ferro2	1	1	2_z	All the elements are allowed: **xxx**, **xyy**, **xzz**, xyz = xzy, xzx = xxz, **xxy** = **xyx**, **yxx**, **yyy**, **yzz**, yyz = yzy, yzx = yxz, **yxy** = **yyx**, zxx, zyy, zzz, **zyz** = **zzy**, **zzx** = **zxz**, zxy = zyx
			$\overline{2}_x$	**xxx**, **xyy**, **xzz**, **xyz** = **xzy**, xzx = xxz, xxy = xyx, yxx, yyy, yzz, yyz = yzy, **yzx** = **yxz**, **yxy** = **yyx**, zxx, zyy, zzz, zyz = zzy, **zzx** = **zxz**, **zxy** = **zyx**
ferro3	2	$1, 2_z$	$\overline{2}_x, \overline{2}_y$	**xyz** = **xzy**, xxz = xzx, yyz = yzy, **yzx** = **yxz**, zxx, zyy, zzz, **zxy** = **zyx**
ferro4	m	$1, \overline{2}_y$	$2_z, \overline{2}_x$	**xxx**, **xyy**, **xzz**, xxz = xzx, yyz = yzy, **yyx** = **yxy**, zxx, zyy, zzz, **zzx** = **zxz**
AF:				
a), g), j)	m	$1, \overline{2}_y$	$2_z, \overline{2}_x$	**xxx**, **xyy**, **xzz**, xxz = xzx, yyz = yzy, **yyx** = **yxy**, zxx, zyy, zzz, **zzx** = **zxz**
b), h), k)	m	$1, \overline{2}_x$	$2_z, \overline{2}_y$	xzx = xxz, **xxy** = **xyx**, **yxx**, **yyy**, **yzz**, yyz = yzy, zxx, zyy, zzz, **zyz** = **zzy**
c), d), l)	2	$1, 2_z$	$\overline{2}_x, \overline{2}_y$	**xyz** = **xzy**, xxz = xzx, yyz = yzy, **yzx** = **yxz**, zxx, zyy, zzz, **zxy** = **zyx**
e), f_a), f_b)	1	1	2_z	All the elements are allowed: **xxx**, **xyy**, **xzz**, xyz = xzy, xzx = xxz, **xxy** = **xyx**, **yxx**, **yyy**, **yzz**, yyz = yzy, yzx = yxz, **yxy** = **yyx**, zxx, zyy, zzz, **zyz** = **zzy**, **zzx** = **zxz**, zxy = zyx
			$\overline{2}_x$	**xxx**, **xyy**, **xzz**, **xyz** = **xzy**, xzx = xxz, xxy = xyx, yxx, yyy, yzz, yyz = yzy, **yzx** = **yxz**, **yxy** = **yyx**, zxx, zyy, zzz, zyz = zzy, **zzx** = **zxz**, **zxy** = **zyx**
i_a), i_b)	mm2	$1, 2_z, \overline{2}_x, \overline{2}_y$	one domain	xxz = xzx, yyz = yzy, zxx, zyy, zzz

TABLE II. Nonvanishing elements of $\chi_{el}^{(2\omega)}$ for all spin configurations of the (111) surface of a fcc lattice. More monolayers are taken into account. We denote $\chi_{ijk}^{(2\omega)}$ by ijk. The configurations are depicted in Fig. 3.

conf.	point grp.	symmetry ops.	domain op.	non-vanishing tensor elements
para	3m	$1, \pm 3_z, \overline{2}_y, \overline{2}_{S(xy)}, \overline{2}_{S(-xy)}$	-	zxx = zyy, xxz = xzx = yyz = yzy, zzz, xxx = -xyy = -yxy = -yyx
ferro1	1	1	$\overline{2}_y$	All the elements are allowed: xxx, xyy, xzz, **xyz** = **xzy**, xzx = xxz, **xxy** = **xyx**, **yxx**, **yyy**, **yzz**, yyz = yzy, **yzx** = **yxz**, yxy = yyx, zxx, zyy, zzz, **zyz** = **zzy**, zzx = zxz, **zxy** = **zyx**
ferro3	m	$1, \overline{2}_y$	-	xxx, xyy, xzz, xxz = xzx, yyz = yzy, yyx = yxy, zxx, zyy, zzz, zzx = zxz
ferro5	3	$1, \pm 3_z$	$\overline{2}_y$	xxx = -xyy = -yxy = -yyx, **xyz** = **xzy** = -**yxz** = -**yzx**, xzx = xxz = yyz = yzy, **xxy** = **xyx** = **yxx** = -**yyy**, zxx = zyy, zzz
AF:				
a_a), a_b), i_a), i_b), k_a), k_b)	m	$1, \overline{2}_y$	-	xxx, xyy, xzz, xxz = xzx, yyz = yzy, yyx = yxy, zxx, zyy, zzz, zzx = zxz
c_a), c_b), f_a), f_b)	1	1	$\overline{2}_y$	All the elements are allowed: xxx, xyy, xzz, **xyz** = **xzy**, xzx = xxz, **xxy** = **xyx**, **yxx**, **yyy**, **yzz**, yyz = yzy, **yzx** = **yxz**, yxy = yyx, zxx, zyy, zzz, **zyz** = **zzy**, zzx = zxz, **zxy** = **zyx**